From: "Seth Putterman"
To: "'wcoblenz''Schmidt, Peter'" "'Adams, Holly'" "'Ken Suslick'"
Subject:
Date: Fri, 6 Apr 2007 15:02:37 -0700

Dear Peter, Bill and Holly!

Attached as a word file [Darpa2phaseReport] is my summary final report for the recent Darpa funded Sonofusion project.
This report includes input from Suslick and Taleyarkhan. Taleyarkhan's complete input to my request for a report is attached as a pdf file
[Report-to-UCLA 4.5.07].

More detail is always available. Please ask if you need something.

Best regards

Seth J. Putterman
310-8252269

Rusi!

In the process of collating the reports the following question came up.
Do you intend to publish a paper regarding the experimental difficulties you encountered trying to reproduce previous work [Science and Phys Rev E] during the period of this project?

Seth

darpa2phaseReport1.doc

Report-to-UCLA_41.5.07_.pdf

SBK: ONR grant N00014-05-1-0459

Final Report regarding the Joint project of Seth Putterman PI, Rusi Taleyarkhan and Ken Suslick to Reproduce Nuclear Fusion in Collapsing Bubbles Surrounded by D-Acetone

SBK: Putterman also submitted it to Holly Adams.

Submitted to Bill Coblenz and Peter Schmidt
April 6, 2007
Submitted by Seth Putterman
310-8252269
puherman@ritva.physics.ucla.edu

SBK: Experiment goal was attempt to replicate 2002 ORNL experiment reported in Science.

SBK: It was not a "carbon copy." There were 6 significant discrepancies.

The goal of this project was to attempt to reproduces Rusi Taleyarkhan's data relating to the observation of fusion inside of collapsing bubbles in Deuterated Acetone. With Taleyarkhan's assistance this project was to attempt to achieve a "carbon copy" of the apparatus that he used to generate data published in Science and Physical Review E.

At a kick-off meeting at Purdue in May 2005 Taleyarkhan presented us with blueprints for his acoustic chamber. This was an important step as Suslick and Putterman had not been given access to this key information prior to the start of this project. Based upon these blueprints UCLA organized for "identical" parts to be produced by various contractors. With the assistance of Steve Hopkins from the lab of Ken Suslick and the Ucla team the experiment was eventually brought on line. As called for by Taleyarkhan's papers [mentioned above] we even purchased [~$80,000.] and brought on line a pulsed neutron generator whose purpose was to seed bubbles into the insonated chamber. From this award and previous DARPA/ONR grants a world class neutron detector was built and merged into this experiment. To the accuracy of our experiment we observed no nuclear fusion with a sensitivity that exceeds that of Taleyarkhan's detector by a factor of 10,000. This work is reported in **Upper Bound for Neutron Emission from Sonoluminescing Bubbles in Deuterated Acetone** ;C. G. Camara, S. D. Hopkins, K. S. Suslick, and S. J. Putterman Phys. Rev. Lett. **98**, 064301 (2007). We made numerous experimental runs in varying arrangements all with null results.

SBK: This is wrong, for many reasons, not the least of which is the fact that if they had the world's best neutron detector, they still wouldn't see anything if they didn't do the experiment correctly. See New Energy Times #31.

In January 2006 Taleyarkhan published a paper in Physical Review Letters [96, 034301 (2006)] which claimed to observe nuclear fusion in bubbles surrounded by an organic liquid. In this case the bubbles were seeded by a dissolved alpha source. Brian Naranjo of Ucla analyzed Taleyarkhan's data and found that it match neutron emission from a 252Cf source and not neutron emission from nuclear fusion. See B. Naranjo; PRL 97, 149403 (2006). Taleyarkhan's reply is on the following page. Taleyarkhan's reply included new data on the response of his detector. Based upon that data Naranjo wrote a comment on Taleyarkhan's reply [http://arxiv.org/abs/physics/0702009] which claims that this new data further substantiates Naranjo's claim regarding the 252Cf source as being the source of Taleyarkhan's data.

SBK: This is Putterman (and Naranjo's) thinly veiled insinuation of fraud. Taleyarkhan responded to Naranjo in PRL. Taleyarkhan disproved Naranjo's computer simulation-based speculation. PRL declined to publish Naranjo's second comment.

On March 1, 2006 the Darpa/ONR team convened at Purdue in Taleyarkhan's lab to observe his acoustic cell in action. The acoustic field was unstable and did not meet Taleyarkhan's published/claimed standards for a sound field that could generate fusion from cavitation.

Putterman twisted this. Taleyarkhan was able to repeat his group's experiment, but only at a 10% success rate because of parts inconsistencies. Regardless, the many failures made the few successes very obvious.

SBK: Putterman creates the appearance of a link between these two paragraphs to suggest in the reader's mind that Taleyarkhan used DARPA funds for the ONR-managed UCLA replication attempt.

Taleyarkhan in his report on this project [see below] says that no publications resulted from his Darpa funded effort at Purdue and that he was unable at Purdue to reproducibly achieve his previously claimed results. These comments refer to his papers in Science[295,1868, (2002)] and Physical Review E [69, 036109 (2004)] wherein neutrons from a pulsed neutron generator were used to seed bubbles into a high sound field.

However, as mentioned above, Taleyarkhan did publish – during the aegis of this project, a paper, wherein he claimed to seed bubbles with a dissolved alpha source. According to this research of Taleyarkhan these bubbles, when acted upon by a sound field expanded and imploded so as to generate nuclear fusion. This paper which appeared in Physical Review Letters [96, 034301 (2006)] was [according to Putterman] substantially funded by proceeds from the award which is the subject of this report. This paper did not acknowledge support from Darpa or any federal agency. Putterman wrote to Taleyarkhan and also to the Purdue administration requesting that an erratum be submitted so as to indicate that this paper was federally funded. Although Putterman claims that the work product which resulted in this paper was substantially funded by the federal government the author and the administration at Purdue have refused to make such an acknowledgment. The paper discussed in this paragraph is the one which was analyzed by Naranjo and shown to yield data consistent with a 252Cf source and not nuclear fusion.

SBK: This is another allegation by Putterman. What he did not know is that this work was not funded by DARPA/ONR and was not part of the ONR grant. Taleyarkhan's 2006 work was funded by the DoE and DHS and funds were not acknowledged in the paper for reasons of national security.

There have always been concerns that the use of volatile organic liquids was not a good route to sonoluminescence or sonofusion. For this reason funds from the Darpa project were used for parallel research on low vapor pressure fluids. In this regard some interesting new science was opened up at Ucla and at the University of Illinois. In Suslick's appended detailed report he discusses [among other contributions] his discovery that sulfuric acid bubbles are extremely bright. At Ucla we measured the dynamical properties of these bubbles [**Dynamics of a Sonoluminescing Bubble in Sulfuric Acid**] Stephen D. Hopkins, Seth J. Putterman, Brian A. Kappus, Kenneth S. Suslick, and Carlos G. Camara Phys. Rev. Lett. **95**, 254301 (2005); as well as contributing to a plasma physics analysis of the high pressures that are developed inside of imploding bubbles **Measurement of Pressure and Density Inside a Single Sonoluminescing Bubble** David J. Flannigan, Stephen D. Hopkins, Carlos G. Camara, Seth J. Putterman, and Kenneth S. Suslick Phys. Rev. Lett. **96**, 204301 (2006). At Ucla we have also been studying a different low vapor pressure fluid- Phosphoric Acid in which case we achieved an upscaling of Sonoluminescence to a peak power of 150Watts. A photo of this line of work appeared in the New York Times February 27, 2007.

SBK: Puttermann and Suslick included text in their proposals to use some of the DARPA/ ONR funds for other research that was of interest to them.

A copy of Taleyarkhan's report regarding this project is attached as a pdf file. As mentioned above he claims to have no results [other than his inability to reproduce previous work] to report regarding this project. I will ask him if he intends to write a paper reporting the experimental difficulties he encountered trying to reproduce his previous work. The slides in his report refer to a presentation which he made at the May 2005 "kickoff" meeting at Purdue. More readable formats are available if desired.

Professor Kenneth S. Suslick
Department of Chemistry, University of Illinois at Urbana-Champaign
600 S. Mathews Av., Urbana, IL 61801.
217-333-2794 ksuslick@uiuc.edu

Overall Technical Objectives

This DARPA project has previously established the importance of chemical reactions during cavitation both in multi-bubble cloud cavitation (MBC) single bubbles (SBC) and their controlling (i.e., limiting) effects on the conditions formed within the collapsing bubble during multibubble sonoluminescence (MBSL) and single bubble sonoluminescence (SBSL). Our understanding of the high energy chemistry that occurs during bubble collapse has allowed us very recently for the first time to generate SBSL in a full series of non-aqueous liquids in air. Two tasks are set for the proposed research to explore the possibility of the creation of neutrons from D-D fusion during the inertial confinement and compression produced by cavitation: (1) exploration of exotic liquids for intensification of acoustic cavitation and (2) the search for neutrons (in collaboration with the Putterman group at UCLA) during MBSL and SBSL in low volatility ("exotic") liquids, including polar aprotic liquids (e.g., N-methylformamide) and non-aqueous *ionic* liquids, at very high acoustic pressures (>10 B).

These projects are part of an ongoing collaboration between this chemical research group at University of Illinois at Urbana-Champaign and Professor Seth Putterman and his group in the Department of Physics at U.C.L.A. This collaborative effort involves a range of expertise unavailable in any one laboratory and includes not only chemical and spectroscopic studies, but also physical acoustics determinations of bubble dynamics and stability regimes, pulse-width measurements of light emission lifetimes, and sophisticated hydrodynamic modeling and simulation of the cavitation process.

Specific Objectives

This project has established the importance of chemical reactions during cavitation both in multi-bubble cloud cavitation (MBC) single bubbles (SBC) and their controlling (i.e., limiting) effects on the conditions formed within the collapsing bubble during multibubble- sonoluminescence (MBSL) and single bubble sonoluminescence (SBSL). Our understanding of the high energy chemistry that occurs during bubble collapse has allowed us recently to generate SBSL in a full series of non-aqueous liquids.

Two tasks (both in collaboration with the Putterman group at UCLA) were set for the proposed research to explore the possibility of the creation of neutrons from D-D fusion during the inertial confinement and compression produced by cavitation: (1) the direct replication of Taleyarkhan's results claiming the production of neutrons and tritium from neutron generated multibubble cavitation in deutero-acetone and (2) the exploration of MBSL and SBSL in "exotic" liquids, including polar aprotic liquids and non-aqueous *ionic* liquids (e.g., sulfuric acid or molten salt eutectics), at very high acoustic pressures (>20 B).

Results

Overall, our research efforts were extraordinarily successful in gaining new and fundamental understandings of acoustic cavitation in non-aqueous liquids. Our DARPA supported studies have led to 11 publications, including 2 papers in *Nature*, 1 communications in *J. Am. Chem. Soc.*, 4 papers in *Physics Review Letters*, and 2 papers in *J. Phys. Chem.*

We were able to dramatically increase the efficiency of conversion of the kinetic energy of cavitation into shock heating of the bubble contents (sometimes referred to as "upscaling") in concentrated sulfuric acid, which produced SBSL that is several thousand times as intense as SBSL in water. There had been previously no strong experimental evidence for even the existence of a plasma during SBSL or multi-bubble sonoluminescence (MBSL). SBSL typically produces featureless emission spectra that reveal little about the intra-cavity physical conditions or chemical processes. Here we present the first definitive evidence of the existence of a hot ionized plasma core during SBSL. We report the first observations of atomic emission (Ar) as well as extensive molecular (SO) and ionic (O_2^+) progressions in SBSL spectra from concentrated $H_2SO_{4(aq)}$. Both Ar and SO emission permit spectroscopic temperature determinations, as accomplished for MBSL with other emitters. The emissive excited states observed from both Ar and O_2^+ are inconsistent with any thermal process: the Ar excited states involved are extremely high in energy (>13 eV) and cannot be thermally populated at the measured Ar emission temperatures (4,000 to >15,000 K); the ionization energy of O_2 is more than twice its bond dissociation energy so O_2^+ likewise cannot be thermally produced. These emitting species must therefore originate from collisions with high energy electrons, ions, or particles from a hot, but optically opaque, plasma core, as is also the case in a star or thermonuclear fireball.

In our most recent work, emission lines from transitions between high-energy states of noble gas atoms (Ne, Ar, Kr, and Xe) and ions (Ar^+, Kr^+, and Xe^+) formed and excited during single-bubble cavitation in sulfuric acid are reported. The excited states responsible for these emission lines range from 8.3 eV (for Xe) to 37.1 eV (for Ar^+) above the respective ground states. Observation of emission lines allows for identification of intra-cavity species responsible for light emission; the populated energy levels indicate the plasma generated during cavitation is comprised of highly energetic particles.

Our attempts in close collaboration with the Putterman group to reproduce Taleyarkhan's claims of inertial confinement fusion through cavitation, however, met with no positive results. In collaboration with the Putterman group (Physics, UCLA), we constructed a resonating cell that is capable of operating at >20 Atm of acoustic pressure without spontaneous cavitation. In addition, as an exact a duplicate of Taleyarkhan's reactor was built.

SBK: Not an exact duplicate.

These were both tested with cavitation induced by a pulsed neutron generator and also by a very low energy laser pulse to seed bubbles in deuterated water or deuterated acetone being driven at approximately 30 kHz.

In our most recent publication in PRL in February 2007 (cf. below for full citation), we report finding ***no*** evidence for excess fusion neutrons in experiments that directly replicate Taleyarkhan's original *Science* paper. This experimental search for nuclear fusion inside imploding bubbles of degassed deuterated acetone at 0°C driven by a 15 atm sound field and seeded with a neutron generator ***revealed an upper bound that is a factor of 10,000 less*** than the signal reported by Taleyarkhan et al. The strength of our upper bound is limited by the weakness of sonoluminescence, which we ascribe to the relatively high vapor pressure of acetone.

Suslick Students Supported with DARPA Funding, 2002-2006:

Yuri Didenko, PDRA, 2002-2005. Now CEO and founder of UT Dots, Inc., Champaign, IL.

Stephen D. Hopkins, Ph.D. student, 2002-2005. Ph.D. Thesis: "Exploring the Limits of Cavitation", January 13, 2005. Now employed by Intel Corp.

David J. Flannigan, Ph.D. student, 2003-2006. Ph.D. Thesis: "Physical Conditions and Chemical Processes During Single-Bubble Sonoluminescence", December 1, 2006. Now a post-doctoral research associate with Professor Ahmed Zewail, California Institute of Technology, Pasadena, CA.

Suslick Papers acknowledging DARPA support (linked to web pdf URLs):

1. Didenko, Y.; Suslick, K. S. "The Energy Efficiency of Formation of Photons, Radicals, and Ions During Single Bubble Cavitation" *Nature* **2002**, *418*, 394-397.
2. Suslick, K. S.; Didenko, Y. T. "The Chemical Consequences of Single-Bubble Cavitation" *Nonlinear Acoustics at the Beginning of the 21st Century*, Rudenko, O.V.; Sapozhnikov, O.A., ed. Moscow State Univ. Press: Moscow, 2002; vol. 2, pp. 1063-1069.
3. Oxley, J. D.; Prozorov, T.; Suslick, K. S. "Sonochemistry and Sonoluminescence of Room-Temperature Ionic Liquids" *J. Am. Chem. Soc.*, **2003**, *125*, 11138-11139.
4. McNamara III, W. B.; Didenko, Y.; Suslick, K. S. "Pressure during Acoustic Cavitation" *J. Phys. Chem.* **2003**, *107*, 7303-7306 (Henglein Festschrift).
5. Flannigan, D. J.; Suslick, K. S. "Plasma Formation and Temperature Measurement during Single-Bubble Cavitation" *Nature*, **2005**, *434*, 52-55.
6. Flannigan, D. J.; Suslick, K. S. "Molecular and atomic emission during single-bubble cavitation in concentrated sulfuric acid" *Acoust. Res. Lett. Online* **2005**, *5*, 157-161.
7. Flannigan, D. J.; Suslick, K. S. "Plasma Line Emission During Single-Bubble Cavitation" *Phys. Rev. Lett.*, **2005**, *95*, 044301-1 - 044301-4.
8. Hopkins, S. D.; Putterman, S. J.; Kappus, B. A.; Suslick, K. S.; Camara, C. G. "Dynamics Of A Sonoluminescing Bubble In Sulfuric Acid" *Phys. Rev. Lett.* **2005**, *95*, 254301-1 - 254301-4.
9. Flannigan, D. J.; Hopkins, S. D.; Camara, C. G.; Putterman, S. J.; Suslick, K. S. "Measurement of Pressure and Density Inside a Single Sonoluminescing Bubble" *Phys. Rev. Lett.* **2006**, *96*, 204301-1 - 204301-4.
10. Flannigan, D. J.; Suslick, K. S. "Plasma Quenching by Air during Single-Bubble Sonoluminescence" *J. Phys. Chem. A*, **2006** *110*, 9315-9318.
11. Camara, C. G.; Hopkins, S. D.; Suslick, K. S.; Putterman, S. J. "Upper Bound for Neutron Emission from Sonoluminescing Bubbles in Deuterated Acetone" *Phys. Rev. Lett.* **2007**, *98*, 064301-1-4.